Report for 2002ME1B: MtBE in Groundwater: The Maine Experiment

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Report Follows:

The Persistence of MtBE in Groundwater in Windham, Maine, USA.

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Abstract

A study was conducted on the occurrence and distribution of the fuel oxygenate MtBE (methyl *tert*-butyl ether) in a glacial sand and gravel aquifer in southern Maine. Ninety samples were collected from 31 different wells in the Windham aquifer, in North Windham, Maine, for analysis of MtBE between July 1998 and August 2001. MtBE was detected in 42 percent of the samples and 52 percent of the individual wells sampled. In addition, 92 percent of wells with detectable concentrations of MtBE were in an area of the aquifer designated as a "high-yielding" aquifer (Neil, 1998). Land uses were found to be associated with MtBE detection rates in the wells in the study area. The overall median concentration in wells with detectable MtBE was 1.85 µg/L. The mean concentration has risen from ~0.5 µg/L in April 1998 to 6.3 µg/L in August 2001. An analysis of the distribution and concentrations of MtBE that were detected in ground water indicates that recharge from precipitation containing MtBE is not a likely source. The strong association of MtBE with land use and aquifer transmissivity suggests a variety of potential land-based sources.

Introduction

The 1992 implementation of the 1990 Clean Air Act mandated oxygenated gasoline containing methyl *tert*-butyl ether (MtBE) in certain areas in the country to meet part of the Federal requirement of reducing VOC concentrations in Maine's air by 15 percent. The State of Maine elected to use reformulated gasoline (RFG) that contained at least 11 percent MtBE by volume (Maine Department of Human Services, 1995). In December 1994, RFG was introduced in southern Maine, in Kennebec, Sagadahoc, Androscoggin, Cumberland, York, Lincoln, and Waldo Counties and by 1995, oxygenated gasoline use was common in the northeast. Soon after the widespread use of gasoline containing MtBE began, water-quality surveys began detecting MtBE in ground water, often without the other sparingly soluble gasoline compounds—benzene, toluene, ethyl benzene, and xylenes (BTEX)—usually found in ground water near gasoline spills (Maine Department of Human Services, 1998).

MtBE has been detected in ground water in Maine as early as 1985. However, it was not until 1998 that evidence of widespread, low-concentration MtBE contamination in the ground water was documented, when several widely publicized instances of contamination in drinking-water wells prompted the State of Maine to conduct a random sampling of 951 private wells and nearly all the 830 non-transient public water supplies in the state of Maine. The sampling found

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MtBE in ground water in all parts of Maine, including areas not required to use RFG (Maine Department of Human Services, 1998). This study and additional studies by the U.S. Geological Survey (USGS) across the country documented low concentrations of MtBE in ground water, especially in urban areas (Zogorski et al., 1998) and in over 8 per cent of public drinking water supplies (Gullick and LeChevallier, 2000). The widespread occurrence of MtBE has prompted discussion about possible nonpoint sources of MtBE in ground water, especially precipitation and atmospheric deposition (Squillace, Pankow et al., 1996; Squillace, Zogorski, et al. 1996; Pankow at al., 1997; Lopes and Bender, 1998; Baehr at al., 1999; Moran et al., 1999). Few studies to date, however, have examined the persistence of MtBE, or how low MtBE concentrations in ground water respond to changes in gasoline formulations.

In 1998, the USGS in cooperation with Senator George J. Mitchell Center at The University of Maine, the Maine Department of Environmental Protection and the Town of Windham, Maine (Figure 1) began a study of MtBE in the Windham aquifer (Nielsen and Peckenham, 2000). The study was designed to collect information on the occurrence and distribution of low concentrations of MtBE (defined as less than 2 µg/L) in the aquifer, a shallow aquifer used for drinking water in an area in which RFG was used exclusively. In this report we evaluate the occurrence of MtBE in groundwater from 1998 during RFG use to 2001, two years after RFG use ceased. Additional sampling and analysis occurred in August 2001. Key objectives are to determine: (1) if MtBE is still detectable in groundwater post-RFG; (2) if the rates of detection and spatial patterns have changed over time; (3) if the spatial patterns relate to potential MtBE sources; and (4) if the concentrations of MtBE change uniformly over time.

Previous Investigations

In 1998, the State of Maine conducted a comprehensive survey of MtBE in ground water, which consisted of sampling 951 randomly selected domestic water wells and 793 of the 830 public supply wells (Maine Department of Human Services, 1998). MtBE was detected at concentrations greater than 0.1 µg/L in 15.8 percent of the residential wells and in 16.0 percent of the public supply wells. More than 90 percent of the detections of MtBE were below 1.0 µg/L. The data were interpreted with respect to well type, knowledge of a recent gasoline spill nearby, population density, and inclusion in the RFG area. No significant differences in MtBE occurrence were observed by well type (drilled bedrock wells compared to unconsolidated surficial wells) or among wells where there were known recent gasoline spills compared to those where there were no known spills (State of Maine, 1998). The State tested the risk of an MtBE occurrence associated with population density and whether or not the well was in a mandatory-RFG area. Both factors were statistically significant, when controlled for the other, in determining the risk of MtBE detections in domestic and public-supply wells. The reason for the smaller percentage of MtBE detections in higher density areas for public-supply wells where RFG was not required was not evident. MtBE detection rates for areas with more than 1,000 people/mi^2 were similar to those for the National Ambient Water Quality Assessment (NAWQA) studies (State of Maine, 1998).

Known and suspected sources of MtBE in ground water include point and non-point sources. Point sources include pipelines, storage tanks (above- and below-ground), accidental spillage, homeowner disposal, spillage during fueling, and waste motor oil. Heating-oil spills

also have been identified as another source of some MtBE in ground water (Robbins et al., 1999). Suspected nonpoint sources include atmospheric deposition (recharge from precipitation or direct transport to ground water through gaseous diffusion), vehicle evaporative losses, and urban runoff (Moran et al., 1999; Baehr et al., 1999).

Point sources will cause locally elevated concentrations of MtBE and other related gasoline compounds. Point sources have the potential to generate very high concentrations of MtBE in water, up to 50,000 mg/L (Barker et al., 1991). The geochemical characteristics of point sources are: local effects with a limited area of contamination, steep concentration gradients with marked changes in concentrations away from source area, and consistent time-series changes in concentrations over time.

Non-point sources, most significantly direct precipitation and subsequent recharge of contaminated rainfall, and stormwater runoff primarily containing gasoline flushed from land surfaces, have been suggested as the source of some low levels of MtBE in ground water (Zogorski et al., 1998; Pankow et al., 1997). Concentrations of MtBE in ground water from non-point sources are limited to low levels, well below 20 µg/L (Moran et al., 1999).

Investigators in a small number of studies have attempted to use models to predict the behavior of aqueous phase MtBE once it starts to infiltrate the unsaturated zone (Johnson et al. 2000). Pankow et al. (1997) modeled various scenarios of recharge in a hypothetical aquifer to test the possibility that MtBE in precipitation could have traveled into shallow ground water during the time frame of its use as a gasoline additive. Assuming no attenuation, they found that it took just 14.3 in/yr of recharge to saturate ground water 6.5 feet below the water table with atmospheric levels of MtBE in 5 years. Baehr et al. (1999) developed a model to predict the concentration of MtBE at the water table, taking into account diffusion, recharge flux, thickness of the unsaturated zone, and several decay functions. They found that the concentration of MtBE at the water table is most sensitive to the thickness of the unsaturated zone and that the reason why MtBE is not universally detected in shallow ground water could be explained by variations in saturated thickness and recharge. Factors that may affect the degradation rate of MtBE, such as aquifer material composition, also would contribute to the heterogeneity of MtBE detection.

MtBE is also present at detectable levels in urban stormwater in areas where RFG was used (Delzer et al., 1996). MtBE was detected in more than 43 percent of stormwater samples collected for permitting requirements for urban stormwater runoff in areas with RFG use. The source of MtBE in the stormwater samples may be either gasoline washoff from paved surfaces or MtBE in the precipitation (Lopes and Bender, 1998). Modeling the behavior of MtBE and other BTEX compounds found in stormwater runoff, at concentrations of MtBE above 1.0 µg/L (the lower detection limit for samples in the study), showed that the source of MtBE was most likely dissolved gasoline entrained during the storm event and not precipitation. This was determined primarily by the co-occurrence of MtBE and other BTEX compounds in the storm-water runoff.

Windham Aquifer Hydrology

The Windham aquifer depicted in Figure 1 is a shallow, glacial sand and gravel aquifer in Cumberland County, southern Maine. It is mapped as a significant sand and gravel aquifer (Neil,

1998), with expected yields ranging from 10 to more than 50 gal/minute (38 to 189 L/m). The aquifer consists primarily of marine deltaic sand and gravel, interbedded in places with a marine silt/clay deposit. A buried esker lies in the northern part of the aquifer (Gerber, Inc., 1997). Thickness of the aquifer ranges from approximately 10 feet to more than 120 feet (3 to 37 m), and depth to water in the aquifer ranges from 5 feet to more than 50 feet (1.5 to 15 m). Hydraulic conductivities calculated from slug tests (Bouwer and Rice, 1976) range from less than 1 foot/day to more than 400 feet/day (0.7 to 122 m/d) (W.J. Nichols, U.S. Geological Survey, written commun., 1999). Average hydraulic conductivities used in a calibrated model of the aquifer (Gerber, Inc., 1997) are 250 feet/day (76 m/d) in the buried esker; 44 feet/day (13 m/d) in the sand and gravel; 1.5 feet/day (0.46) in the till surrounding the aquifer; and less than 0.1 feet/day (0.03 m/d) in the marine silt/clay. Flow paths based on observed and modeled heads indicate recharge areas in the western and southwestern areas of the aquifer and discharge areas at Outlet Brook and on the edges of the aquifer (Gerber, Inc., 1997; W.J. Nichols, written commun., 1999). Some recharge also may come from Little Sebago Lake, a heavily developed lake that borders the Windham aquifer on the northeast.

Temperature data are available for Portland, Maine, 14 miles (22.4 km) southeast of Windham and the average annual temperature at Portland is 45.4°F (7.4°C) (Northeast Climate Center, 2002). Temperatures were consistently greater than the mean for 1998 (48.3°F/9.0°C), 1999 (47.9°/8.8°C), and 2001 (47.0°F/8.3°C). Average annual precipitation near Windham is 44.34 inches (1.12 m). Precipitation varied markedly from the mean for the study years 1998 (54.77 inches/ 1.39 m), 1999 (40.70 inches/1.03 m), and 2001 (32.93 inches/0.84 m).

The potential for MtBE-bearing precipitation to recharge the Windham aquifer depends on the amount of precipitation, amount of ground-water recharge, and average yearly and monthly temperatures. An estimate of the maximum amount of yearly recharge to sand and gravel aquifers can be calculated using average annual temperature to estimate evapotranspiration and precipitation (Lyford and Cohen, 1988). The estimated average annual temperature of 43°F (6.1°C) translates into an annual evapotranspiration of 19.5 inches (0.49 m). This evapotranspiration combined with an annual average precipitation of 44 inches (1.12 m) for the Windham area, yields a maximum potential recharge to the Windham aquifer of 22 in/yr (0.56 m). In the Windham area, some runoff is collected in detention basins that route runoff from paved areas into ground-water recharge zones and some is surface runoff.

Methods

Ground-water samples

An existing network of shallow wells (Figure 1), installed by the USGS and distributed throughout the ground-water flow system in the Windham aquifer was used for this study (Nichols and Silverman, 1998). These wells are completed in different types of glacial sediments. Samples were collected in July and August 1998 in 31 wells; subsequent sampling rounds in November and December 1998, April and May 1999, and August 2001, included a smaller number of wells (Table 1). Well depths ranged from 19 to 132 feet (5.8 to 40 m), with screened interval depths ranging from 8 to 130 feet (2.4 to 39.6 m). Screened intervals were mostly 5 or 10 feet (1.5 to 3.0 m) long, with a few wells having longer screens, up to 30 feet (9.1 m). Depth to water in the wells at the time of sampling ranged from 4.5 to 61.1 feet (1.37 to 18.6

m). All the wells are completed in unconfined parts of the aquifer; the sampling points range from water-table position to more than 110 feet (33.5 m) below the water table. Seven pairs of nested wells were sampled to evaluate vertical distributions of MtBE. The dominant land cover (Table 1) near each well was determined from the NLCD land-cover classification: urban, undeveloped, or low-density residential**.**

USGS National Water Quality Assessment protocols for sampling ground-water wells (Koterba et al., 1995) were followed in sample handling, quality assurance/quality control (QA/QC), sampling equipment, and cleaning. The USGS protocols were modified, however, to follow the U.S. Environmental Protection Agency (USEPA) low-flow (minimal drawdown) sampling procedures (Puls and Barcelona, 1995). A small number of wells did not yield enough water to meet the minimum drawdown requirements. These wells were pumped dry and sampled the next day.

The sampling equipment setup was simplified from USGS protocols to reflect what was necessary for the small number of constituents sampled in this study. A submersible Fultz 300R pump with Teflon-lined tubing was used to collect samples. The Teflon outflow tube was connected to a Hydrolab flow-through cell for monitoring field parameters. Once field parameters stabilized as defined as a change of less than 5 percent for three consecutive measurements and drawdown was kept to within 20 percent of the standing head, the flow rate was measured and the sample collected. Flow rates were generally within the range of 0.026 to 0.11 gal/min (100 mL/min to 400 mL/min). Samples for analysis of VOCs (benzene, toluene, ethyl benzene, xylene, and methyl tert-butyl ether) were collected directly from the Teflon outflow tube into three 40-mL amber glass volatile organic analysis vials with septum tops and no air space. The collection method was modified in 2001 to minimize cross-contamination by using disposal polyethylene bailers following the purging of the wells. Samples were kept on ice and shipped within 24 hours to the USGS laboratory for analysis for the 1998 and 1999 samples and the Mitchell Center laboratory for the 2001 samples. After sampling, all the equipment was cleaned with a dilute Liquinox solution, rinsed with distilled water, rinsed with methanol, and rinsed again with distilled water. The pump and cleaned tubing were packed in plastic sheeting for transport between sites.

Quality assurance samples consisted of equipment blanks (15 percent of all samples), trip blanks, spiked samples, source-solution blanks, and ambient blanks (trip blank vials opened and exposed to the ambient air during sampling, then closed and shipped for analysis). In all, 26 percent of all the samples analyzed were QA/QC samples. During the initial round of sampling, deionized water created in the District laboratory was used for rinsing; this presented a problem, however, in that the water was later found to be contaminated with very low concentrations of VOCs. An additional round of samples was collected for all the environmental samples with VOC detections in the first round to eliminate the rinse water as a source of VOCs to the samples. Purchased distilled water was used for all additional sampling, and all sampling equipment and supplies were stored in a clean environment.

Detections of MtBE in the first round of samples (11 detections) were recoded as a “less than” value larger than the observed analytical result. For example, a detection in the first round of 0.42 µg/L would have been recoded to <0.5 µg/L, to make sure that the database did not

contain detections of MtBE from potentially compromised samples. MtBE was also detected in subsequent sampling rounds in 9 of the 11 wells with in which it was detected in the first round. Statistical analyses were performed using SYSTAT ver. 9.

PERSISTENCE OF MTBE IN THE NORTH WINDHAM AREA

Analyses of the ground-water samples conclusively demonstrate that MtBE is widely distributed at low concentrations in the Windham aquifer. Additionally, time-series results indicate that MtBE is persistent and widespread throughout the aquifer.

MtBE in ground water

MtBE was detected in the majority of the water samples from the Windham aquifer. This result is based on analyses of 90 individual samples collected from 31 wells (Table 1). A cumulative total of one to four water samples were collected from each of the wells on different sampling dates in 1998, 1999, and 2001. Sixteen of the 31 wells sampled (52 percent) had at least one detection of MtBE (Table 1). MtBE was detected in 38 individual samples (42 percent of all samples), and the median concentration of MtBE was 1.85 µg/L (excluding the samples that were recoded as "less thans" from the first sampling round). MtBE was below the detection limit in 52 samples (58 percent); however, 13 of these samples had elevated detection limits at greater than 0.2 µg/L. Two of the three highest concentrations detected, 14.0 µg/L and 6.64 µg/L, were collected from the same well, CW 2012. This well is located in an urbanized area, close to a gasoline station. The single highest concentration, 38.7 µg/L, was detected in well CW 2004 located in a light-industrial and commercial area.

The detection of MtBE was accompanied by the detection of the other sparingly soluble BTEX compounds- benzene, or toluene in less than 10 percent of the samples. This observation is consistent with other findings of MtBE along with some BTEX in trace concentrations in Maine (State of Maine, 1998). Since 1998 the median concentration for detections has varied from 0.35 µg/L in 1998 to 2.17 µg/L in Winter 1998 to 0.50 µg/L in 2001 (Figure 2). Over the same period the mean MtBE concentration has increased consistently from 0.50 to 6.30 µg/L. This trend has been driven by a few high concentrations from the same group of three wells (CW 2004, CW 2005, and CW 2012).

To determine if there were any spatial patterns to the MtBE detections, the data were analyzed with respect to: depth of the sample within the aquifer; location of sample in either the high- or moderate-yielding parts of the aquifer as mapped by the Maine Geological Survey (Neil, 1998); and land cover. In addition, the association of MtBE with hydraulic conductivity (K) independent of aquifer yield zone was analyzed (Nielsen and Peckenham, 2000).

Fourteen of the wells sampled were paired wells screened in both the shallow and deep parts of the aquifer. Using the distribution of detections in the paired wells, a Chi-square test (Ott, 1993) failed to detect a significant difference between the concentrations of MtBE in shallow and deep parts of the aquifer. The probability of detecting MtBE is nearly equal in each well of a pair. An analysis of the entire sample population with respect to depth of sampling

point shows a higher number of detections in the shallow parts of the aquifer as defined as <50 feet (15.2 m) deep, but there are still enough detections at depth to affect statistical significance.

To test the effect of aquifer transmissivity, the spatial distribution of MtBE detections was grouped according to location in the aquifer as either high-yield zone (i.e. wells are expected to produce more than 50 gal/min) or moderate-yield zone (i.e. wells are expected to produce between 10 and 50 gal/min), as mapped by the Maine Geological Survey (Neil, 1998). The high-yield zone had many more wells with detections (92 percent) compared to the moderate-yield zone (58 percent). These two subgroups are significantly different ($p = 0.05$). In this report, p-values less than 0.05 indicate that the null hypothesis of the test was rejected.

The wells in which MtBE was detected were grouped according to the land-cover classification defined as: undeveloped, urban, and low-density residential (Figure 2). Although the concentration of detected MtBE was not noticeably different among the groups, the frequency of MtBE detection was quite different. MtBE was detected in 83 percent of the samples collected from wells in the low-density residential area, in 50 percent of samples from the urban area, and in 60 percent of samples from the undeveloped areas. A Kruskall-Wallis test (Helsel and Hirsch, 1992) on the MtBE concentrations by land-cover classification showed that at least one group is significantly different from the others ($p = 0.013$) when compared by pairs. The significance level varied by collection date with the samples collected in the winter of 1998 exhibiting the greatest level of significance.

MtBE detections are more frequent in the areas of the aquifer with higher K values than in areas with lower K values. Hydraulic conductivity for each well, as determined by slug tests (W.J. Nichols, written commun., 1999) was compared to MtBE detections for wells having at least one detection per well. For this test, the wells were divided into two equal groups of K ranges: < 5 feet/day and > 5 feet/day. A 2x2 contingency table test (Chi-square test) shows a statistically significant difference ($p=0.049$) in the distribution of wells with detections in the two groups.

SUMMARY AND CONCLUSIONS

A study was conducted on the occurrence and distribution of the fuel oxygenate MtBE (methyl *tert*-butyl ether) in a glacial sand and gravel aquifer in southern Maine. Ninety samples were collected from 31 different wells in the Windham aquifer, in North Windham, Maine, for analysis of MtBE between July 1998 and August 2001. MtBE was detected in 42 percent of the samples and 52 percent of the individual wells sampled. In addition, 92 percent of wells with detectable concentrations of MtBE were in an area of the aquifer designated as a “high-yielding” aquifer (Neil, 1998). MtBE was much more commonly detected in wells with measured hydraulic conductivities more than 5 feet/d (1.5 m/d) than in wells with hydraulic conductivities less than 5 feet/d (1.5 m/d). Urban and low-density residential land uses were found to be associated with MtBE in the wells in the study area. The median concentration in wells with detectable MtBE was 1.85 µg/L.

Before Maine opted out of the RFG program in March 1999, median concentrations of MtBE in air at the North Windham site were 0.25 ppbv (Nielsen and Peckenham, 2000). According to this study, after Maine stopped using RFG, the median concentration in air dropped to 0.09 ppbv, but no MtBE was detected in the precipitation samples obtained after RFG phase-out. On the basis of the equilibrium partitioning of MtBE from the air into rain, the predicted average concentration of MtBE in rainfall during months when recharge typically takes place (October-December and March-April) is approximately 0.3 to 0.4 µg/L when Maine was using RFG and approximately 0.1 µg/L after the phaseout of RFG. The concentration before the phaseout would be detectable in ground water if the recharge reached the water table with no degradation of MtBE. After the phaseout, it is unlikely that MtBE from recharge would be detectable in ground water.

An analysis of the distribution and concentrations of MtBE that were detected in ground water indicates that recharge from precipitation containing MtBE is not a likely explanation for the occurrence of MtBE in the Windham aquifer. The mean concentration of MtBE detections in ground water has increased from ~0.5 µg/L in April 1998 to 6.3 µg/L in August 2001, while the median has been near 0.5 µg/L since Spring 1999. The strong association of MtBE with land use and aquifer transmissivity/hydraulic conductivity suggests land-based sources. The mechanisms by which MtBE enters the aquifer were not identified in this study. The Maine State study of MtBE tentatively identified very small spills of gasoline associated with filling of lawn-care equipment fuel tanks, recreational vehicle tanks, and automobile tanks as likely being responsible for the majority of the low levels of MtBE found. If true, MtBE should be detectable statewide in similar urban to suburban settings.

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Figure 1. Location of Windham Aquifer and Groundwater Wells. Aquifer boundaries from Neil (1988), map based on Nielsen and Peckenham (2000).

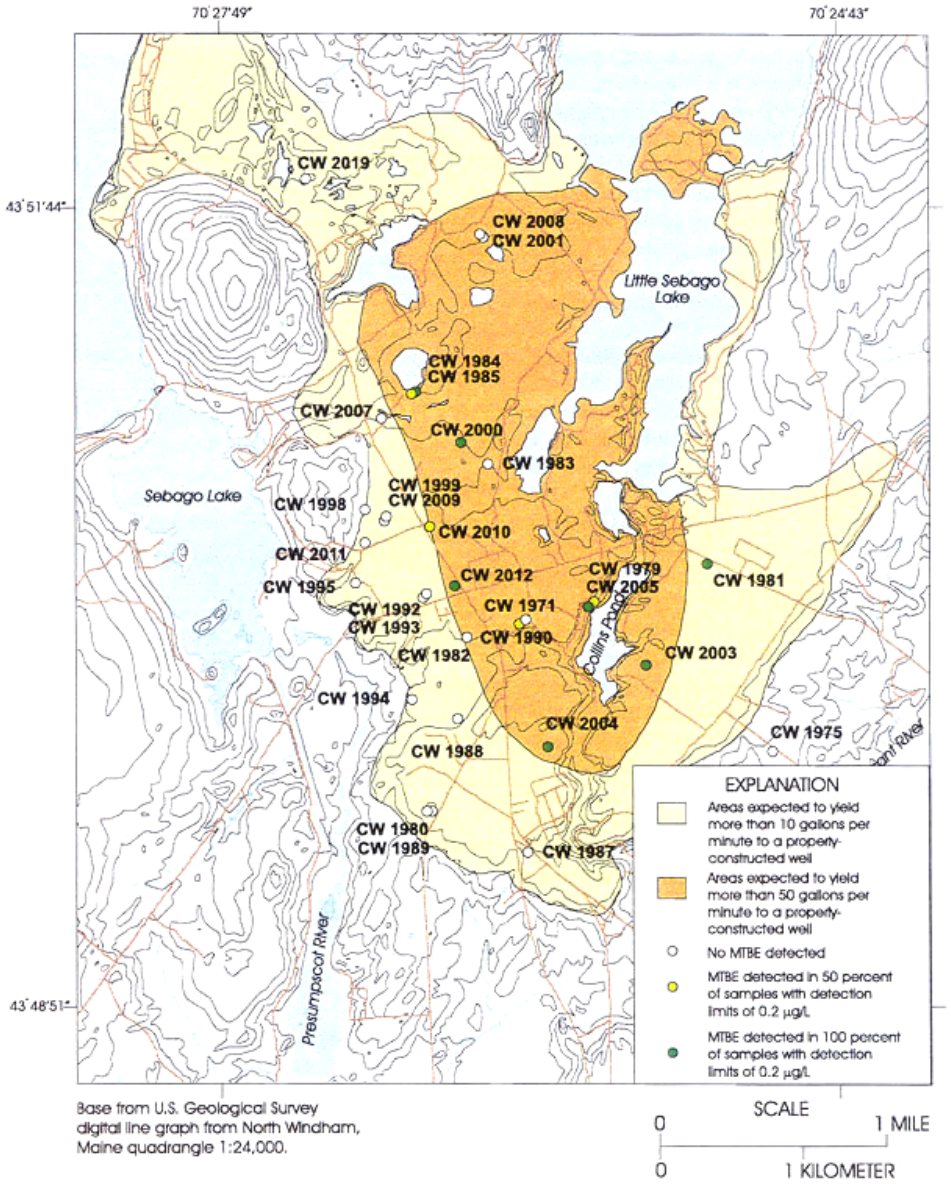

Figure 2. MtBE Concentrations Summaries by Sample Date and Land Use Category.

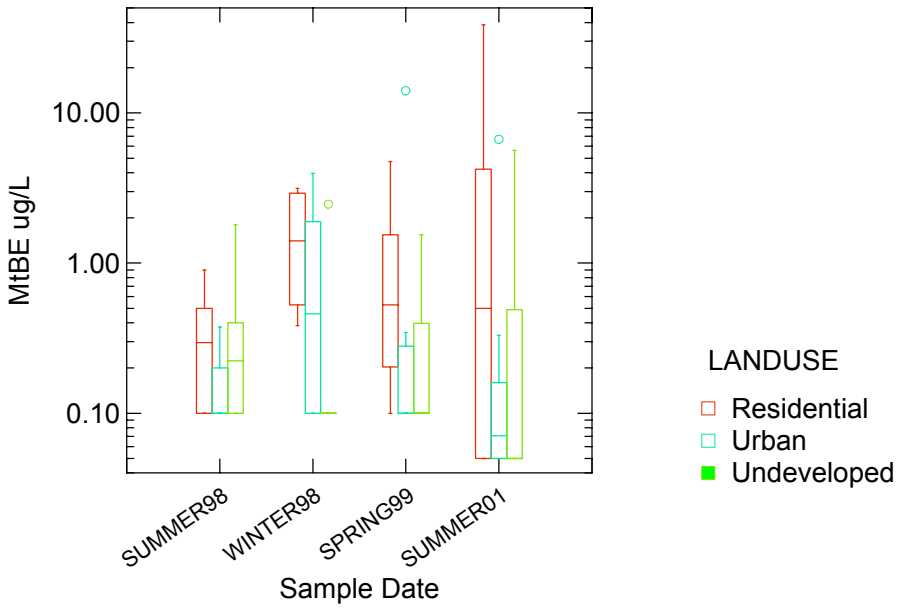

Table I. Summary of Sample Locations and Analytical Results.

Station Identification		Land Use[1]	Sample Date and MtBE Detections (ug/L)[2].			
Station	**Couplet**		**Summer 98**	**Winter 98**	**Spring 99**	**Summer 01**
CW 1971	CW 1990	U	N		N	N
CW 1975		Z	N			
CW 1979	CW 2005	L	N		**0.20**	**0.50**
CW 1980	CW 1989	Z	N			**0.49**
CW 1981		L	**<0.25**	**0.30**	**0.47**	
CW 1982		L	N			
CW 1983		Z	N			
CW 1984	CW 1985	Z	**<1.80**	**2.46**	**1.54**	
CW 1985	CW 1984	Z	**<0.25**	N	**0.10**	N
CW 1987		L	N		N	N
CW 1988		L	**<0.30**			
CW 1989	CW 1980	Z	N			
CW 1990	CW 1971	U	N		**0.23**	N
CW 1992	CW 1993	U	N	N		N
CW 1993	CW 1992	U	N		N	N
CW 1994		Z	N			
CW 1995		Z	N	N		
CW 1998		Z	**<0.35**		N	**0.10**
CW 1999	CW 2009	U	N		N	N
CW 2000		U	**<0.20**	**0.46**	**0.35**	**0.16**
CW 2001	CW 2008	Z	**<0.40**	N	N	N
CW 2003		L	**<0.50**	**0.72**	**0.47**	N
CW 2004		L	**<0.50**	**3.14**	**0.6**	**38.7**
CW 2005	CW 1979	L	**<0.90**	**2.73**	**1.54**	**4.22**
CW 2007		L	N			
CW 2008	CW 2001	Z	N	N		**5.65**
CW 2009	CW 1999	U	N	N		N
CW 2010		U	N	**1.89**		**0.33**
CW 2011		Z	N	N	N	N
CW 2012		U	**0.37**	**3.96**	**14**	**6.64**
CW 2019		Z	N			
	n=	31	31	16	21	22

Note 1. Land Use Codes: L = Residential, U = Urban, and Z = Undeveloped.
Note 2. Detections only are reported. N = None Detected, Blank = No Sample, < = see text for explanation.